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SENSITIVE LASER SPECTROSCOPY MEASUREMENT TECHNIQUE. (U)

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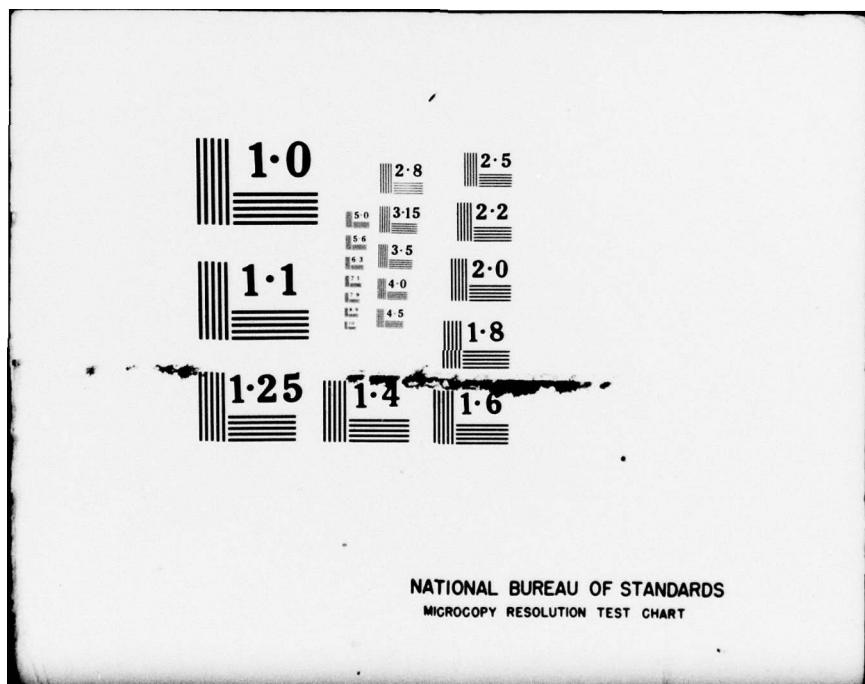
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## Sensitive Laser Spectroscopy Measurement Technique

Aerophysics Laboratory  
The Ivan A. Getting Laboratories  
The Aerospace Corporation  
El Segundo, Calif. 90245

Interim Report

28 September 1977

Prepared for  
**AIR FORCE WEAPONS LABORATORY**  
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This report has been reviewed by the Information Office (OI) and is releasable to the National Technical Information Service (NTIS). At NTIS, it will be available to the general public, including foreign nations.

This technical report has been reviewed and is approved for publication.  
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20. ABSTRACT (Continue on reverse side if necessary and identify by block number)	<p>A simple differential measurement technique is described by means of which a sensitive determination can be made of a change in the laser-beam signal resulting from absorption, scattering, or gain while traversing a medium. The method depends upon the use of an internally modulatable laser for implementation. Sensitivities of <math>\Delta I/I_0 = 10^{-4}</math> were routinely and unambiguously measured. Maximum sensitivity was measured at <math>2 \times 10^{-5}</math>. The method is applicable, in general, to steady phenomena or average value measurements. Two silicon photovoltaic detectors are employed in the differential measurement.</p>		

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The applicable wavelength domain of this technique is determined by the sensitivity range of the detectors, i.e., approximately 0.19 to 1.15  $\mu$ m.

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## I. INTRODUCTION

A sensitive  $F_2$  absorption diagnostic technique was recently developed for measuring low-level  $F_2$  concentrations in supersonic jets of continuous-wave HF or DF chemical lasers. Laboratory sensitivities were demonstrated to  $\Delta I/I_0 = 10^{-4}$  and then were further improved to  $2 \times 10^{-5}$ . This sensitive absorption diagnostic technique was developed for a specific application but has promise in laser spectroscopy experimentation in the ultraviolet, visible, and near infrared wavelengths.

## II. METHOD

In the system are used a laser radiation source and a dual beam and detection scheme that allow the differential measurement of small changes in the probe-beam intensity, relative to the reference-beam intensity (Fig. 1). Several system requirements must be met to attain measurement sensitivities of the order of  $10^{-4}$  to  $10^{-5}$ .

The radiation source must be a laser with an output power of 1 mW or greater. This constraint is imposed by the sensitivity of the silicon photo-voltaic detectors. The detector wavelength sensitivity range also constrains the laser wavelengths from approximately 0.19 to 1.15  $\mu\text{m}$ . The laser must be internally modulatable, with a 100% modulation depth. The usual practice of providing mechanical chopping of the beam results in the introduction of differential transient light-beam signals at the instants of on- and off-beam switching in the two optical paths, causing a sensitivity limitation of the order of  $10^{-2}$  to  $2 \times 10^{-3}$ , e.g., as with absorption spectrophotometers. Two internal modulation schemes were investigated, i.e., acousto-optic intra-cavity modulation and plasma modulation. These methods of modulation produce undistorted beams colinear with the optic axis during the modulation process, minimizing or eliminating the switching transients caused by a scanning of the reference and probe beams over different inhomogeneous optical surfaces during chopping in mechanical systems. It is expected that dye-laser modulation by means of pump-laser modulation will also produce undistorted beams. This contention, however, has not been demonstrated and will be the subject of further study.

Intensity differences in the two light-path beams of the optical system may develop as a result of polarization effects. Since reflectivity and transmissivity of the optical components in the optical paths are functions of the incident-beam polarization, independent intensity changes in the two polarization components of an unpolarized laser beam may produce uncompensated

beam-intensity changes. Hence, a prism is placed in the optical path before beam splitting to eliminate one of the beam polarizations. A thick optical dielectric flat or an optical wedge is used as the beam splitter for ease of elimination of one of the reflected beams to avoid noise generation in the reference-beam detector as a result of interference. The angle of orientation of the flat, relative to the incident beam, is nominally 45 deg but may be varied within limits to help achieve a closer balance in the reference- and probe-beam intensities at the detectors. An uncoated dielectric flat is used when the probe-beam detector requires use of a narrow-band optical transmission filter with reduced in-band transmission, which reduces the transmitted beam of the beam splitter and brings it into closer balance with the reflected beam. If no optical filter is required, a dielectric-coated mirror (~45% transmission, 45% reflectivity) may be used.

The beam of higher intensity also passes through a suitable linear optical density wedge of limited opacity range, e.g., quartz, 0 to 0.3. Translation of this optical density wedge transverse to the optical path reduces the refined intensity of the stronger beam to bring the two beams measured at the detectors into the condition of zero differential signal. Suitable, high-quality optical flats, e.g.,  $\text{CaF}_2$  or quartz, can be used as absorption-region windows. The beam must be free of any limiting apertures in the optical path from laser to detectors. In addition, the optical path lengths to the two detectors should be about the same to ensure nearly equal spot sizes. Sensitive laser spectroscopy measurements were made with the system shown in Fig. 1 with the use of silicon photovoltaic EG&G Model UV 444B detectors. Attempts to use photomultiplier tubes were unsuccessful. Hence, the method is currently limited by the spectral response curve of the detectors (Fig. 2).

The detectors were connected to a Tektronix Model 1A7A 10- $\mu\text{V}$  sensitivity differential amplifier and viewed on a Model 545 oscilloscope. The load resistors were  $1000\ \Omega$  each. These load resistors maintained photovoltaic operation in the linear region of the characteristic curve. The detector radiation power sensitivity is nominally of constant value (within

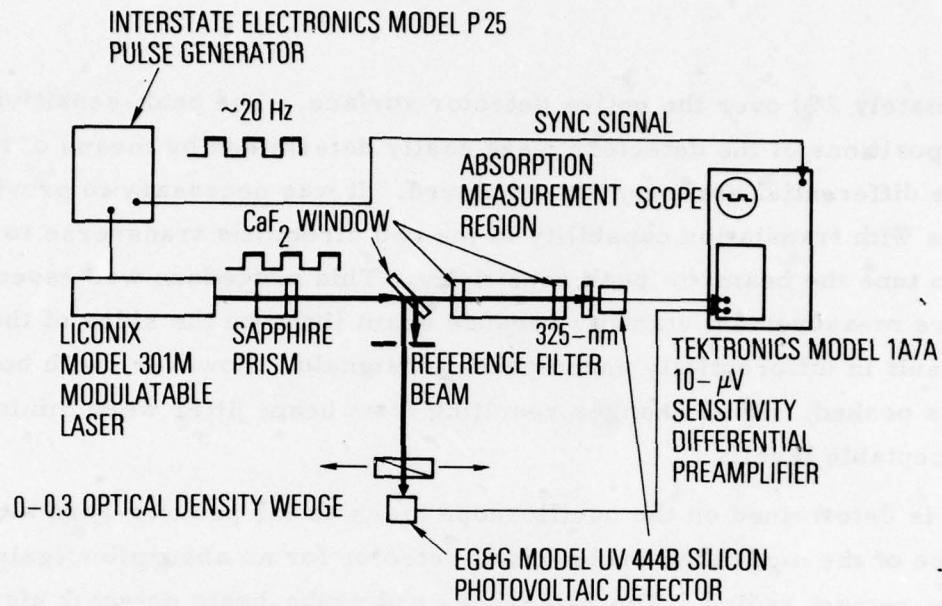


Fig. 1. Sensitive Laser Spectroscopy Measurement Technique

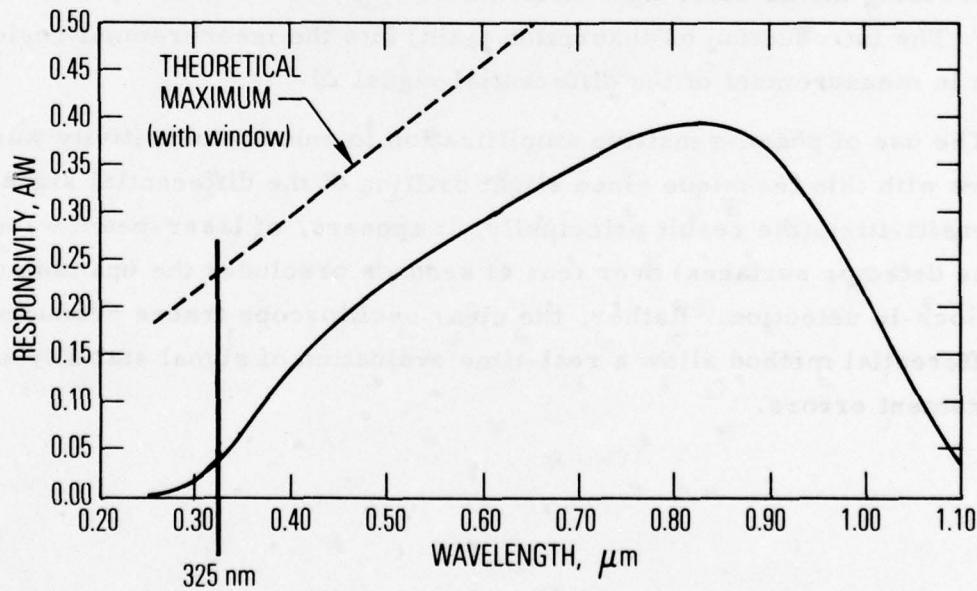


Fig. 2. Typical Silicon Photovoltaic Detector Spectral Response Curve

approximately 2%) over the entire detector surface. The peak-sensitivity surface positions of the detectors were easily determined by means of the sensitive differential arrangement employed. It was necessary to provide the detectors with translation capability in the two directions transverse to the beams to tune the beams to peak sensitivity. This procedure was essential to achieve measurement stability because beam jitter on the sides of the peak could result in differentially uncompensated signals. However, with both detectors peaked, signal changes resulting from beam jitter were minimized to an acceptable level.

$I_0$  is determined on the oscilloscope trace as the peak-to-zero signal difference of the modulated probe-beam detector for no absorption (gain) in the measurement region. The reference- and probe-beam detector signals are brought into coincidence (differential oscilloscope signal = 0 within  $10^{-4}$  to  $10^{-5}$ ) by viewing the difference signal at decreasing amplifier gain levels while reducing the detector light differential by positioning the optical density wedge. The introduction of absorption (gain) into the measurement region results in measurement of the differential signal  $\Delta I$ .

The use of phase-sensitive amplification to enhance sensitivity was not effective with this technique since slight drifting of the differential signal at high sensitivities (the result principally, it appears, of laser-beam wander over the detector surfaces) over tens of seconds precludes the unambiguous use of lock-in detection. Rather, the clear oscilloscope traces produced by this differential method allow a real-time evaluation of signal stability and measurement errors.

### III. EXPERIMENTAL DEMONSTRATION

The sensitive laser spectroscopy measurement technique was demonstrated at 325.0- and 441.6-nm wavelengths by means of square-wave, acousto-optically modulated HeCd lasers (Liconix Models 301M and 405M) at beam power levels of 1, 10, and 15 mW. This type of modulation was most effective. The square-wave rise and fall times were 700 and 200 nsec.

Oscilloscope traces of the laboratory data obtained with the Liconix Model 301M modulatable HeCd laser operating at 325.0 nm and 1 mV are shown in Fig. 3. Chopping frequency for these traces is nominally 18 Hz for each case. The reference (-B) and probe (A) signals as viewed independently on the oscilloscope are shown in superposition in Fig. 3(a). The signal level, corresponding to  $I_o$ , is 12 mV and is displayed on either side of the zero (no light) level. Note the absence of any transient effects at the instants of chopping, either on or off. The top trace in Fig. 3(b) is the -B trace alone with a reduced amplifier frequency bandpass (0 to 1 kHz), relative to the display in Fig. 3(a) (0 to 1 MHz). Simultaneous display of the balanced A - B signals results in the middle trace in Fig. 3(b). These traces are both observed at a 10-mV/cm differential gain. The bottom trace in this figure is displayed at a gain three orders of magnitude smaller than the two top traces, i.e., 10  $\mu$ V/cm, where the previously unobservable imbalance of 9  $\mu$ V ( $\Delta I/I_o = 8 \times 10^{-4}$ ) is seen. Note the almost total elimination of laser signal noise. Further refinements in a signal presentation are shown in Figs. 3(c) and 3(d). The amplifier bandpass was reduced to 100 Hz, Fig. 3(d). A lower limit sensitivity in 1 part of  $10^{-4}$  is shown by the trace in the middle.

Plasma modulation of a 1-mW HeNe laser operating at 632.8 nm was also employed to successfully demonstrate this technique. The laser was powered by a half-wave, rectified electrical power supply. For this type of operation, the  $I_o$  and differential signals were measured from the no-signal levels to the peaks of the half-wave laser signals, Figs. 4(a) and 4(b).

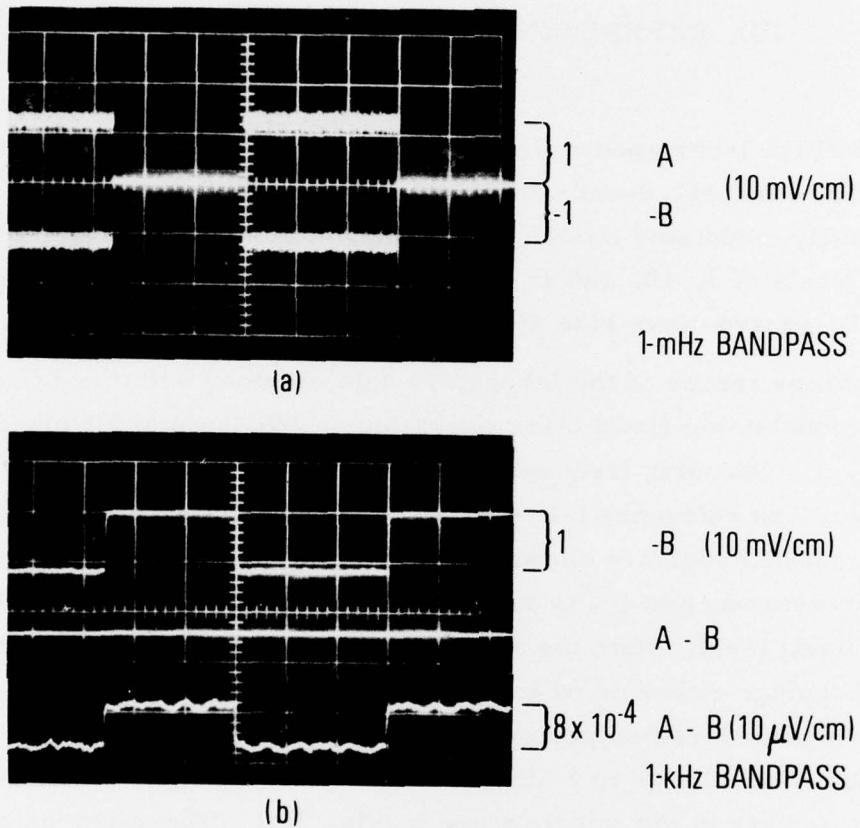


Fig. 3. 325-nm Laser Differential Measurement Scope Traces ( $10^{-4}$  Sensitivity). (a) and (b).

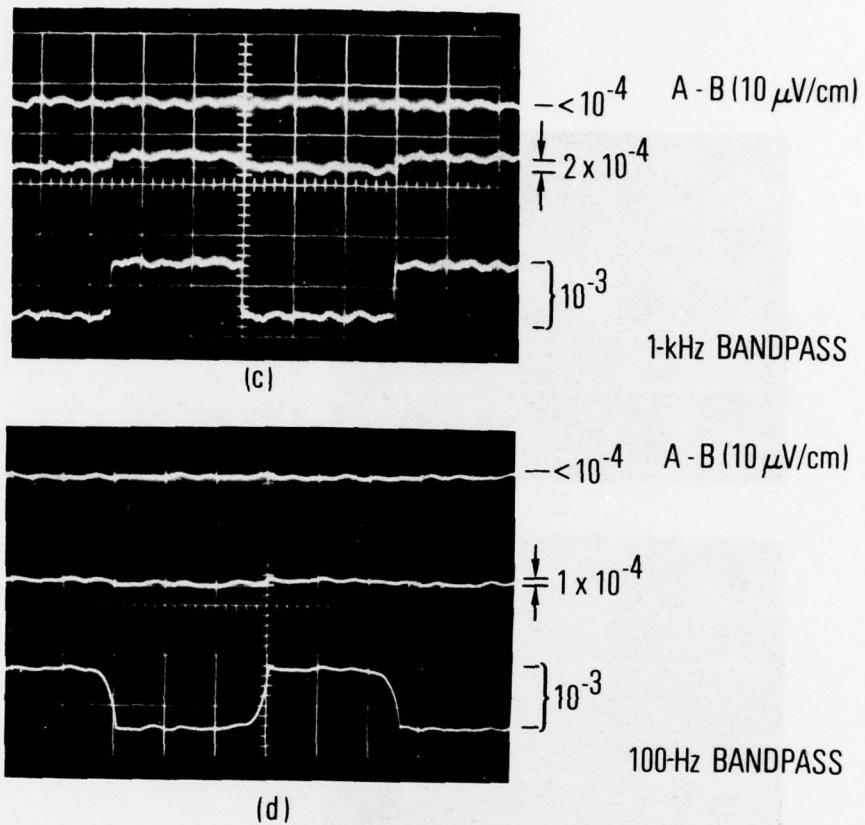
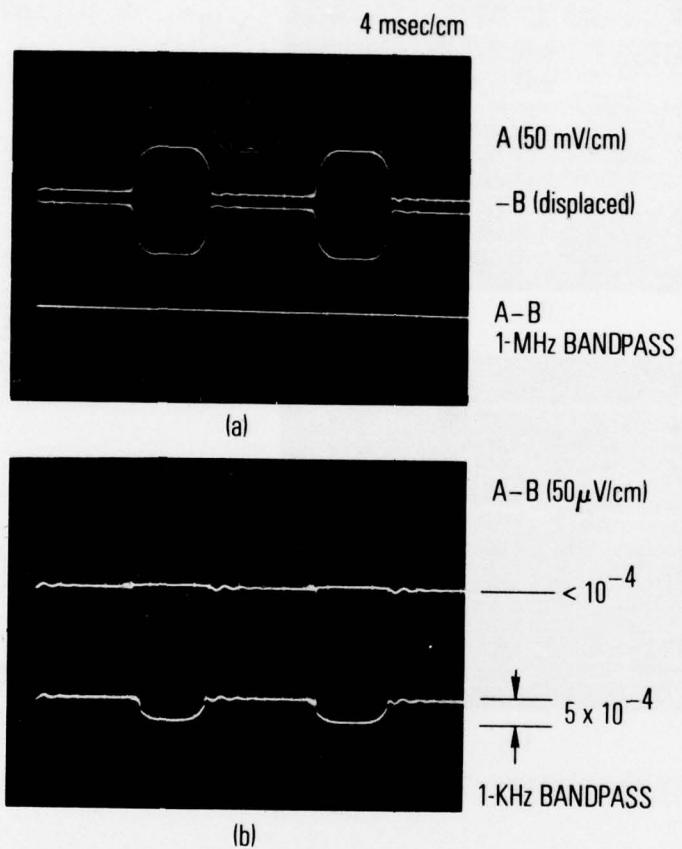


Fig. 3. 325-nm Laser Differential Measurement Scope Traces ( $10^{-4}$  Sensitivity). (c) and (d).



**Fig. 4. Half-Wave, Rectified Power, Modulated HeNe Laser Differential Measurement Scope Traces**

The method was successfully applied directly to the measurement of low-density  $F_2$  concentrations in a cold-flow calibration facility and the supersonic jets of HF and DF continuous-wave chemical lasers. For these experiments, a 325-nm wavelength operation is required. This wavelength is near an insensitive ultraviolet end of the detector response curve, hence, represents a somewhat difficult portion of the operational spectral range (Fig. 2).

This system, when used as an  $F_2$  absorption diagnostic, was calibrated directly in an  $F_2$  cold-flow facility.  $F_2$  density along a path length between windows was determined by introducing  $F_2$  in an  $N_2$  diluent at known partial pressures into the flowing measurement chamber while monitoring steady-state chamber pressure and temperature. A flowing system was used to eliminate  $F_2$  wall-loss effects. Excellent agreement (within plus or minus 2%) was obtained over the range  $\Delta I/I_0 = 8 \times 10^{-4}$  to  $2.9 \times 10^{-2}$  between the measured absorption and the calculated absorption on the basis of  $F_2$  density (Fig. 5).

A higher powered HeCd laser (Liconix Model 405M) operating at 15 mW was used to demonstrate this method, and increased sensitivity was obtained. The experimental setup is shown in Fig. 6. The detectors are housed in Cu blocks to reduce thermal drift. Some difficulty was experienced with this laser. The square-wave lasing signal was marred by an overshoot at the beginning of the pulse, resulting in a differential scope presentation that was not as perfect as the signals observed with the Model 301M laser. Nevertheless, increased sensitivity,  $\Delta I/I_0 = 2 \times 10^{-5}$ , was obtained (Fig. 7). At this sensitivity level, the principal difficulty in measuring is in long-term drift, which in a period of 10 to 20 sec will shift the differential reading by  $2 \times 10^{-5}$ . This change in differential reading is thought to be the result of beam wander.

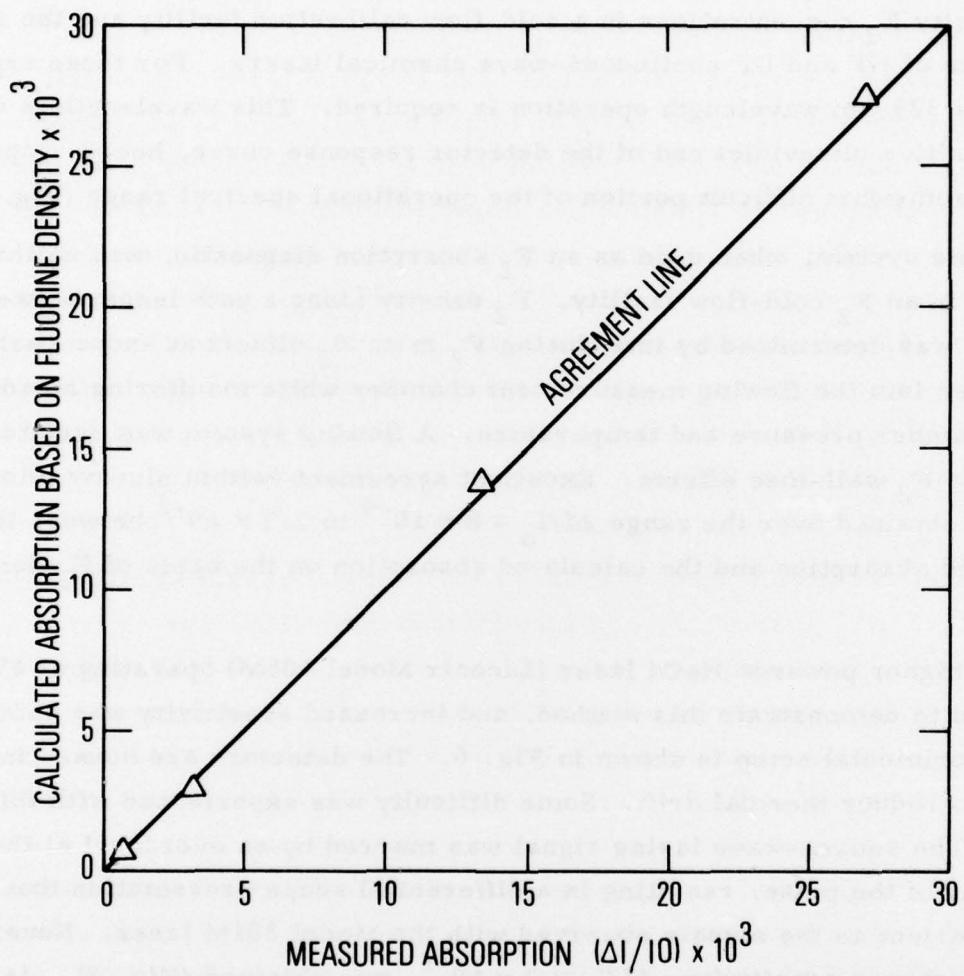


Fig. 5.  $F_2$  Absorption Calibration in Cold-Flow Facility

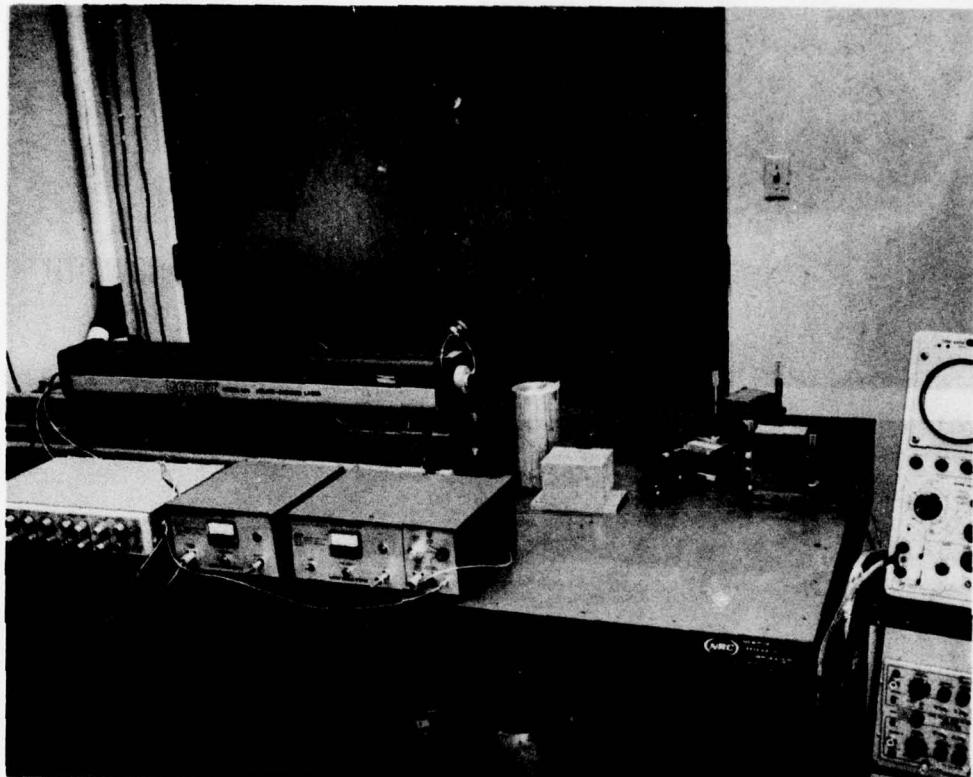


Fig. 6. Differential Measurement Experimental Array

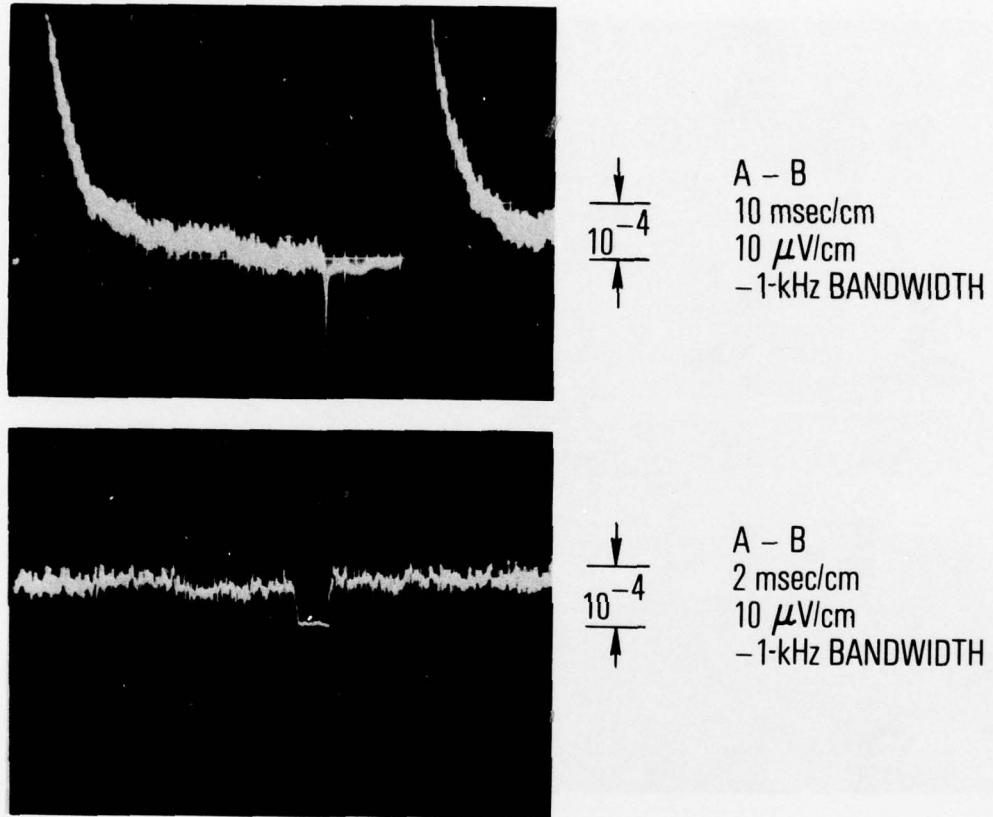


Fig. 7. 325-nm Laser Differential Measurement Scope Traces  
( $2 \times 10^{-5}$  Sensitivity)

#### IV. DYE-LASER APPLICATIONS

The demonstration of this technique at 325 nm, near the insensitive short wavelength tail of the silicon-photovoltaic-detector response curve, implies equally good (or better) system performance in the region of greater detector sensitivity at longer wavelengths because of the inherently higher detector signal-to-noise ratios. Performance was demonstrated at both the 441.6- and 632.8-nm wavelengths. Application of this method to the current continuous-wave dye-laser spectral range (approximately 400 to 950 nm) should be relatively straightforward. In particular, a dye laser employing the folded, three-mirror, astigmatically compensated resonator with non-collinear laser pumping appears to lend itself readily to this measurement technique (Fig. 8)[3]. The pump beam is focused on the dye jet stream with spot sizes of the order of 10  $\mu\text{m}$ . Because of the dye laser threshold, modulation of the pump laser to a 100% modulation depth is unnecessary. Plasma modulation of the pump laser to a reduced power level, rather than complete discharge turn off, eliminates restart problems and laser-beam transients. Simple mechanical chopping of the pump laser beam or of a focused pump-laser beam may be adequate for implementation of this technique. Investigation of this sensitive measurement method in which are used a Coherent Radiation Model CR-3000K SG krypton ion pump laser and a Model CR 599-21 dye laser [3] with a folded, three-mirror, jetstream configuration is contemplated for the near future.

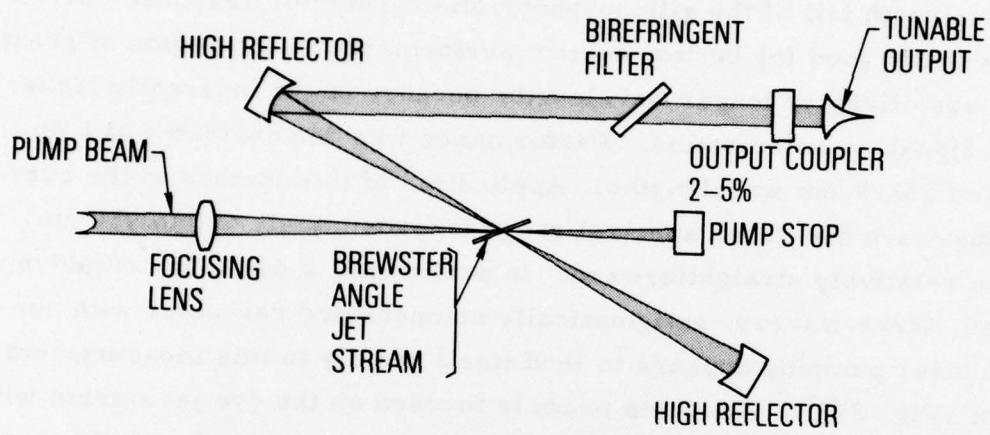


Fig. 8. Noncollinear Pumped Dye Laser

## V. TRANSIENT PHENOMENA

This technique was developed for the study of steady-state phenomena or average-value measurements. Chopping frequencies of order 20 to 60 Hz were used. High sensitivity requires high-frequency electronic filtering of all frequencies greater than 1 kHz (Fig. 3). The detector rise time is of the order of  $20 \times 10^{-9}$  sec. Hence, all limiting time constants associated with the detector in this application are derived from the associated circuitry. The cabling to the oscilloscope provides the limiting-circuit time constant and is easily maintained at  $< 10^{-6}$  sec. The measurement of transient changes of this time order, e.g., in flowing systems, with this system requires a relaxation of the electronic-filtering, high-frequency bandpass, hence, a lower achievable sensitivity. Transient phenomena of approximately 1  $\mu$ sec are observable at sensitivities of approximately  $10^{-3}$ .

## VI. CONCLUDING REMARKS

The differential measurement technique was demonstrated at sensitivities to  $2 \times 10^{-5}$ , an improvement of 2 orders of magnitude over mechanical chopping differential measurement techniques currently employed in many laboratories. The chopping frequencies used to date with this technique range from approximately 20 to 60 Hz. Electronic filtering of all frequencies greater than 1 kHz is also employed. Hence, the method is generally applicable to steady phenomena or average value measurements. However, transient phenomena of approximately 1  $\mu$ sec are observable at sensitivities of  $10^{-3}$ . It is expected that this technique could be used for all internally modulatable laser systems operating within the detector sensitivity range. In particular, plasma modulation (or, perhaps, even mechanical chopping) of the pump laser in dye-laser systems will make this increased sensitive laser spectroscopy measurement technique applicable at all wavelengths provided by the current dye-laser systems.

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